Decadal effects of solid industrial wastes on the coastal environment: Gulf of Gabes (Tunisia, Southern Mediterranean Sea) as an example

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Abstract

The present work examines the different types of solid effluents dumped in the coastal environment from Gabes phosphate fertilizer plants, describes their coastal repartition and assesses their long-term environmental impacts. Scanning Electron Microscopy analysis of collected beach sediment samples showed that these wastes consist of phosphogypsum, fluorapatite (phosphate ore grains), smokey quartz, calcite, fluorite, calcite-Mg, apatite, pyrite, albite, sphalerite-Cd-U, fluorosilicates, sulfur dross and spent catalysts. Using fluorapatite as a mineralogical marker of the coastal spatial distribution of industrial wastes, solid effluents were found to carry bottom and/or suspended loads with a coastal transport limited at Essorrag Wadi, ~ 10 km south from the industrial emissary. Phosphogypsum which is the main solid effluent was found to be completely dissolved in the seawater. The main decadal consequences of solid effluents on the coast were found to be the following: local disappearance of *Posidonia oceanica* seagrass, formation of an azoic benthic zone between Gabes and Ghannouche ports, gradual fattening of the beach, shoreline advancement, formation of a belt of coastal dunes, mouth clogging of coastal streams and formation of coastal lakes.

Keywords: Anthropogenic pressure; Solid wastes; Coastal modifications; Tunisian Chemical Group; Phosphates; Fluorapatite.

1. Introduction

Due to the increasing urbanization and industrialization of the coasts, huge quantities of industrial/solid wastes end in the marine environment, leading to its deterioration (Tosic et al., 2018) and consequently to various socio-economic impacts. For example, it is estimated that the Mediterranean Sea receives yearly 650 10^6 t of sewages, 129 10^3 t of mineral oil, 60 10^3 t of mercury, 36 10^3 t of phosphates and 3.8 10^2 t of lead (UNEP, 2012). In addition to the environmental and health issues related to solid waste disposal in the marine environment, solid wastes can be responsible for a range of economic impacts on marine and coastal activities reducing the economic benefits derived from them (Newman et al., 2015). In fact, in the absence of a good management strategy, solid wastes, in particular industrial ones, can impact the fragile coastal ecosystems, economic sectors such as tourism and fisheries and even public health (Kindt, 1984; Alam and Ahmade, 2013; Groeneveld et al., 2018). Industrial solid wastes are of particular concern because they may consist of various harmful pollutants including heavy metals and radioelements which can deteriorate the marine life and flora leading to the contamination, erosion and modification of the coast. Among the various industrial solid wastes dumped in the Mediterranean marine environment, phosphogypsum (PG; Periáñez, 2002) is an industrial radiochemical by-product of phosphoric acid production which is environmentally hazardous and potentially harmful to human health (El Zrelli et al., 2018a).

The Gulf of Gabes (GG) extends from Ras Kaboudia in the north to the Tunisian-Libyan borders in the south (Fig. 1). It represents the widest continental shelf area in the Mediterranean Basin, together with the North-Central Adriatic shelf, covering about 36.10^3 km^2 (Hattab et al., 2013; Halouani et al., 2016). The Gulf of Gabes is known to have very large continental plateau with a very light slope, making the 200m isobath reachable at almost 250 km from the coastline (Seurat, 1934; Hattour and Ben Mustapha, 2013). GG had been considered as one of the most productive Mediterranean regions (Abdou et al., 2016) and one of the most important fishing zones (Papaconstantinou and Farrugio, 2000), with a variety of fishing activities (Rabaoui et al., 2010; Hattab et al., 2011), due mainly to the largest area covered by *Posidonia oceanica* seagrass beds that it used to host (De Gaillande, 1970; Batisse and De Grissac, 1998; Hattour and Ben Mustapha, 2013). In spite of its various and important ecological and economic services, the GG is currently considered as the most heavily polluted Mediterranean marine area (El Zrelli et al., 2017). In particular, in its central part, various anthropogenic factors are continuously exerting pressure on its natural habitats making them

very vulnerable. For instance, the central GG coastal area is the destination of various industrial and domestic pollutants and wastes which have immensely contributed to the destruction of its specifics flora (e.g. *Posidonia oceanica*; Zaouali, 1993; El Zrelli et al., 2017), fauna (Darmoul and Vitiello, 1980; Rabaoui et al., 2010, 2015; Ayadi et al., 2016; El Kateb et al., 2016), as well as its sediments (Ayadi et al., 2014; El Zrelli et al., 2015; Rabaoui et al., 2015; El Kateb et al., 2018) and seawater quality (Darmoul et al., 1980; Darmoul, 1988; El Zrelli et al., 2018b; El Kateb et al., 2018). Among these different industrial wastes, solid industrial effluents which are mainly represented by PG by-produced from the Tunisian Chemical Group of Gabes (Groupe Chimique Tunisien, GCT), raise a major environmental concern for the coastal habitats of central GG (Darmoul, 1988; Rabaoui et al., 2014; El Zrelli et al., 2015; El Kateb et al., 2016). In fact, a daily average of ~30 10^3 tons of untreated humid PG (El Zrelli et al., 2017) and other unknown industrial wastes have been continuously thrown, from the GCT in the central GG coastal area. Theses wastes are responsible of a clear damage of the local marine environment, biodiversity loss and modification of the coast, since the first observations of Darmoul et al. in 1976 (Darmoul et al., 1980). Thus, it is very probable that these wastes have affected, along the last decades, the coastal landscape of central GG at different levels. The present study was conducted for the first time within this context and it aims at *i*) identifying the different types of solid industrial effluents discharged in the marine environment, *ii*) describing their costal spatial distribution and *iii*) assessing their long-term effects on this area.

2. Materials and Methods

2.1. Sampling area, trenching and beach sediment sampling

In September/October 2014, 12 sedimentary coastal trenches were made at 10 sampling stations spread at the transitional zone between the wrack line and the beach berm, between the marine discharge of GCT (source) and Zarat Beach (Fig. 1). At the beginning of the study, only one trenche was realized at each station, but after the examination of the trenches done, we decided to make 2 additional trenches in Chat Essalam in order to better understand and confirm the distinguished sedimentary profile in this latter station. At each station, trenching was aborted when reaching the water level, and hence the depth of the trenches varied from 0.65 (at Tebelbou) to > 2.20 m (at Chat Essalam). Systematically, after digging each trench, one composite sediment sample (made by mixing three different samples) was collected from each sediment layer observed in each trenche. In total, 23 composite sediment samples were

collected with a rate of only 1 sample from each of the trenches done in LM, KT and ZT, 2 samples from each of the trenches in LG, CR, OG, CM, MT and TB, and 2 to 3 samples from each of the trenches dug in Chat Essalam (i.e. CE1, CE2 and CE3) (Table 1; Fig. 1).

The study area was divided into 2 sectors:

- Inter-harbour Sector: located between the commercial harbor of Ghannouche in the north and the fishing port of Gabes city in the south (Fig. 1). It extends along 2.5 km between Legusir Wadi which is used as the industrial releases emissary of GCT and Ennaten Wadi to the south (Fig. 1). It contains 2 stations (LG and CE) and is characterised by relatively low hydrodynamic energy conditions (Gzam et al., 2016) due of the influence of the structures of the two ports (El Zrelli et al., 2015).
- Southern Sector: located toward the south and is characterized by moderate energy conditions (Gzam et al., 2016). It extends nearly along 30 km from the fishing port of Gabes to Zarat Beach in the south (Fig. 1) This sector hosts the other 8 sampling stations: CR, OG, CM, MT, TB, LM, KT and ZT (Fig. 1).

In addition to the 23 sediment samples collected from the ten sampling stations above-mentioned, 2 additional sediment samples were taken directly from the littoral industrial emissary of GCT. The first sample (E1) was taken at the level of the industrial canal bed where the current is strong, in order to have the coarse fraction of the dumped industrial wastes. However, the second sample (E2) is much finer because it comes from the delta of industrial channel where the environment is less agitated (low hydrodynamics energy). These 2 additional samples were collected in order to identify the different types of industrial wastes thrown from the coastal phosphate fertilizer factories of Gabes city. Collected sediment samples were placed separately in sealed and clean polyethylene bags. In the laboratory, samples were dried in an oven at 60°C for few days to a constant weight.

2.2. Mineralogical, morphological and granulometric characterization of beach sediments and solid wastes.

The collected sediment samples were first observed under a binocular loupe in order to determine their general characteristics (color, size, shape, etc.). After embedding the sediment samples, polished sections were prepared and observed under a Scanning Electron Microscopy (SEM; JEOL JSM 6360LV) coupled with an Energy Dispersive Spectroscopy detector (EDS; SAHARA Silicon Drift Detector). The SEM-EDS analysis enabled to

166 determine the mineralogical, morphological and granulometric characteristics of each
167 sediment sample.
168
169 *2.3. Sedimentological characterization of the beach area in the two sectors*
170 Each of the two sectors defined herein was chosen to serve a well-defined purpose. The inter-
171 harbour sector was firstly used (*i*) to identify the different types of industrial wastes thrown
172 from the GCT factories because it shelters in its northern part the littoral channel of these
173 wastes. This sector is limited by the piers of the commercial and fishing harbours which trap a
174 large part of industrial wastes, making it the most affected area by these industrial discharges
175 (Ayadi et al., 2014; El Zrelli et al., 2015). Hence, this sector was defined (*ii*) to identify the
176 impacts of phosphate plant discharges on the coastal environment. Sediment samples
177 collected from the different layers of the trenches done in Chat Essalam station, were used to
178 assess the concentration of the inorganic phosphorus (P_{ing}), cadmium (Cd), Zinc (Zn),
179 Chromium (Cr), Copper (Cu), Mercury (Hg) and Lead (Pb). Elemental analyses were
180 conducted following the methodology described in El Zrelli et al. (2015).
181 Because of its location in the direction of PG marine dispersion (El Zrelli et al., 2015) and
182 hence in the direction of discharged solid wastes, the southern sector represents the ideal area
183 (*iii*) to determine the spatial limit of GCT solid wastes' sedimentary transport in central GG.
184
185 *2.4. Spatial limitation of the coastal transport of industrial solid wastes*
186 In order to spatially limit the littoral extension of sedimentary transport of GCT solid wastes,
187 the fluorapatite, one of the main insoluble minerals of phosphate ore grains dumped by the
188 coastal factories into the seawater of GG (present study), was used as a "mineralogical
189 marker" of industrial waste littoral transport. The choice of fluorapatite is also confirmed with
190 the study of Poizat (1970) which was conducted, before the setup of the phosphate fertilizer
191 plant (in 1972), on the analysis of GG coastal sediments and did not report the presence of this
192 mineral. For this purpose, we have used the optical and electronic microscopy to minutely
193 search the presence of this marker in the sediments from the southern zone in the direction of
194 transport of these industrial wastes (El Zrelli et al., 2015). We started with the southernmost
195 station (Zarat) towards the industrial wastes source. The principle of the method used is too
196 simple: the first southern station, where fluorapatite is found, represents the limit of industrial
197 wastes transport.
198

2.5. *Assessment of the long-term effects of PG industrial wastes on the central coastal area of GG*

After several years of observation (1990-2018) and during many field campaigns carried out in our study area (2013-2017), several field surveys were done with local habitants in order to determine the coastal modifications since the beginning of solid wastes dumping in the littoral environment (1972). For the same purpose, we have studied and compared several ancient photos (since 1952), aerial (since 1949) and satellite images from different free sources (since 1984 from Google Earth and LandSat) of central part of GG.

3. Results and discussion

3.1. Qualitative identification of different types of industrial solid wastes

The SEM observation of two sediment samples collected from the bed (E1, coarse fraction) and delta (E2, fine fraction) of the marine effluents, from the industrial discharges of Gabes phosphate chemical complex, showed the presence of several types of minerals. The coarse fraction (Fig. 2) consisted essentially of fluorapatite, smokey quartz, calcite, fluorite, sulfur dross, spent catalysts (i.e. solid wastes produced during the process of sulfuric acid production and composed mainly by C, O and Si), sphalerite-Cd-U (Fig. 3), apatite and fluorosilicates. The fine fraction was found to be formed by phosphogypsum, smokey quartz, fluorapatite, calcite-Mg, sphalerite-Cd, fluorite, pyrite and albite. The diversity of identified minerals showed that GCT plants do not only throw PG wastes (~30.10^3 t/day of humid PG; El Zrelli et al., 2017), but also phosphate ore grains and other untreated industrial solid wastes like sulfur dross and used catalysts.

Historically speaking, the first studies on the environmental impacts of the GCT industrial wastes on the Gabes Gulf reported liquid effluents as the only form of industrial wastes discharged in the marine environment at the beginning of coastal industry (Darmoul et al., 1980a and b; Darmoul, 1988). Along the decades following the setup of the phosphate industry in Gabes coast, the industrial wastes have been intensified and diversified. This has been accentuated with the absence of priorities of marine environment protection in the seventies and development of GCT industries, leading to increase the anthropogenic pressure on the coastal environment which has become the receiving area in which end all generated wastes. Compared with other coastal areas hosting phosphate industries such as Sfax (Tunisia; Gargouri et al., 2010; Naifar et al., 2018), Huelva (Spain; Morillo et al., 2004; Pérez-López et al., 2011), El Jadida (Morocco; Gaudry et al., 2007), Kpémé (Togo; Bawa et al., 2006) and

Batroun (Lebanon; Fakhri et al., 2008), the observation of industrial solid wastes, including phosphate ores, spent catalysts and sulfur dross, being discharged in Gabes coastal waters was reported only during this study.

3.2. Sedimentological characterization of different beach layers

The examination of the twelve beach trenches carried out in the studied area allowed to distinguish 4 successive layers, L1, L2, L3 and L4, in the inter-harbour area and only one contaminated layer (L_S) in southern sector. They are sedimentologically and mineralogically heterogeneous (Figs. 4 & 5; Table 2). The L1 layer was found throughout the study area (Zone 1, 2 and 3; Fig. 5) and consisted of the regional white fine-grained sand and was found to be mineralogically uncontaminated with the presence of quartz, calcite and some opaque grains essentially represented by zircon, iron oxides and titanomagnetite minerals (Table 2). The L2, L3 and L4 layers are exclusively located in the inter-harbor area (Fig. 4; Zone 1 in Fig. 5) and were found to be characterized by coarser particle size and darker color than the bottom layer L1 (Table 2). In addition, these last 3 layers are contaminated because of their enrichment with several minerals of industrial origin such as fluorapatite, calcite-Mg, sphalerite-Cd-U, fluorosilicates and some sulfur dross (SM-Fig. 1); these latter were not found in L1 (see more details in Table 2). Besides, it is also important to note that L4 layer could be considered as a typical industrial wastes layer, formed by almost only phosphate ore grains (Table 2). Indeed, even with naked eyes, this layer was found to be distinguished from the other two "mineralogically" contaminated layers (L2 and L3) by the dominance of phosphate raw minerals, mainly fluorapatite, which gave it the dark brown to blackish color. Finally, the Ls layer is spatially limited in Zone 2 of the southern sector (Fig. 5) and is composed by fine-sized sand grains and characterized with a greyish color, due to the presence of calcite-Mg (Table 2).

Fig. 5 shows the East-West (E-W) and North-South (N-S) interpretative diagrams of the trench profiles found in our study area, showing the sequential organization of the different layers already defined in Fig. 4 and described in Table 2.

The E-W section of Zone 1 (Fig. 5A) showed the occurrence of 3 main different phases of industrial discharges (corresponding to the three layers L2, L3 and L4), since the set-up of the phosphate fertilizer industry along the coast of Gabes (in 1972). These three layers are simultaneously marked with their horizontal increase of grain size and their contents of phosphate grains, in particular fluorapatite, from the base (L2) to the surface (L4). These

granulometric and mineralogical differences are related to the changes in industrial processes and consequently the changes in the types of industrial wastes released. Within this context, it is worth noting that the last industrial phase which corresponds to the surface layer, L4, was found to be granulometrically, mineralogically and quantitatively the most important compared to the two others phases (L2 and L3). Indeed, this last phase is the main phase responsible for the various radical morphological changes seen at present, in particular in the inter-harbor beach (Zone 1; Fig. 5; SM-Fig. 2). Figure 6 showed the variations of elements (P_{ing}, Cd, Zn, Cr, Cu, Hg and Pb) concentrations analyzed in the sediment samples collected from Chatt Essalam Beach layers. The concentrations of P_{ing} varied between 0.15 g kg^{-1} (in L1) and 39.1 g kg^{-1} (in L3); those of Cd between 4 mg kg^{-1} (in L1) and 723 mg kg^{-1} (in L4). The concentrations of Cr were found to vary from 10 mg kg^{-1} (in L1) to 57 mg kg^{-1} (in L4). In the case of Cu, concentrations ranged between 2.2 mg kg^{-1} (in L1) and 6.4 mg kg^{-1} (in L4). Regarding the concentrations of Hg, they varied from 0.011 mg kg^{-1} (in L1) and 0.026 mg kg^{-1} (in L4). As for Pb, its concentrations ranged between 4.8 mg kg^{-1} (in L1) and 11.9 mg kg^{-1} (in L2); those of Zn between 4.9 mg kg^{-1} (in L1) and 4732 mg kg^{-1} (in L4). In addition, the lowest concentrations of all elements taken into consideration were recorded in the uncontaminated layer L1; whereas the highest concentrations were found in the other three layers (L2, L3 and L4; Fig. 3). The geochemical enrichment of these three latter (upper) layers is most likely due to their 'mineralogical contamination' by the typical phosphate minerals mainly fluorapatite and sphalerite-Cd (Sassi and Sassi, 1999).

As for the N-S section profile (Fig. 5D), it showed that the industrial impacts on the beach environment are not only concentrated in the inter-harbor zone (Fig. 5A and D) but they are also extended to southward-spreading (Zone 2; Fig.5B), with a very much lower amplitude. A thin (up to 80 cm) dark-greyish contaminated layer (L_s) is found to overlay the regional uncontaminated white sands. It ends at Essorrag Wadi (Fig. 5D) thus marking the spatial limit of coastal solid waste transport at this level (~10 km from the source).

Comparing between known phosphate industries in the Mediterranean Sea, the largest marine releases of phosphogypsum are being dumped by the GCT plants of Gabes (~30 10^3 tons of untreated humid PG; El Zrelli et al., 2017). For example, in Batroun region (North Lebanon), only 950 t of PG are daily discharged in the sea (METAP/Tebodin, 1998) which represents more than 30 times less than the quantities rejected in Gabes. In addition, the other insoluble industrial wastes such as phosphate ores, sulfur dross and used catalysts are discharged only in the coastal waters. All other phosphate industries do not follow the same practices adopted

by Gabes phosphate factories. These latter fractions (i.e. phosphate ores, sulfur dross and used catalysts) cannot be dissolved in the seawater end are therefore washed on the coastal beaches where they accumulate with years to form stratified layers with respect to the type of wastes they form.

3.3. Spatial limitation of the solid industrial wastes impact on the central coastal area of Gabes Gulf: Spatial repartition of fluorapatite mineral

Fluorapatite grains, the main mineral component of Tunisian phosphate ores, were used as the mineralogical marker of the coastal spatial distribution of solid GCT wastes, since they predominantly occur in all the contaminated layers of the inter-harbor (L2 to L4) and southern (Corniche to Tebelbou sampling stations, L_s) sectors). Figure 7 showed the scanning electron microscopy photos of some representative fluorapatite mineral grains from 3 stations: Legusir (LG; the source of GCT solid wastes), Chatt Essalam (CE; the most impacted coastal zone) and Tebelbou (TB; the limit of the presence of contaminated industrial layer L_s). These grains are distinguished by different shapes and sizes, with respect to the sampling stations. The LG fluorapatite grains (Fig. 7) are characterized by elongated shapes and large sizes (between 1050 and 1200 µm). However, in the two other stations (CE and TB), the fluorapatite grains have almost the same spherical shape but with different sizes: CE fluorapatite grains ranging between 600 and 800 µm are larger than those of TB fluorapatite grains (between 230 and 280µm). Within this context, each grain size may be associated with a specific mechanism of transport (Wilcock et al., 2009) in a moderate hydrodynamic context like the study area of GG (Maghrebi, 1995; Guillaumont et al., 1995; Hattour et al., 2010). Industrial emissary (LG) fluorapatite is transported only in bed load by sliding and rolling or hopping according to Wilcock et al. (2009). Two transport mechanisms are possible for the inter-harour sector fluorapatite grains (Chatt Essalam): bed load and suspended load. Finally, the fluorapatite mineral grains of the southern sector move exclusively in suspension with the seawater mass along the southern-directed littoral drift (suspended load; Wilcock et al., 2009). This transport mode may be added to another particular mode: the transport in colloidal suspension by the PG foams (SM-Fig. 3). According to El Zrelli et al. (2019), PG foams can migrate far into the marine environment (reaching tens of km) transporting with them small grains of fluorapatite and depositing them after their mechanical collapse as shown in SM-Fig. 3. These results showed that the sedimentary coastal transport of GCT solid wastes - essentially phosphates grains – is limited spatially at the level of Essorrag Wadi, at ~10 km far from their source.

Besides, it is worth noting that from Legusir to Zarat stations (~30 km), no trace of PG, the main solid wastes of phosphoric acid factories, was detected except for the area located in front of the industrial discharge delta. Hence, it is strongly suggested that PG is completely dissolved in the seawater. This dissolution contributes enormously to the marine dispersion of the polluting load of these radiochemical industrial wastes (El Zrelli et al., 2018a).

In other areas hosting phosphate industries such as Batroun (Lebanon), El Jadida (Morocco) and Seine estuary (France; where PG discharges stopped in 1992), the environmental impacts of the PG industrial wastes remain spatially limited (Fakhri et al., 2009, 2011; Simon et al., 2000), contrary to what is being observed in Gabes. Various anthropogenic and natural factors can explain the current status of the coastal area in GG. For instance, the important quantities (~$10.5.10^6$ t/year of humid PG; El Zrelli et al., 2017) and diversity of industrial discharges (Fig. 2), spatial extension and low inclination of the continental shelf (Hattab et al., 2013; Halouani et al., 2016; Hattour and Ben Mustapha, 2013) and the role played by harbors' marine structures (Rabaoui et al., 2014; El Zrelli et al., 2015) act in synergy in order to enlarge the spatial extent of the environmental impacts of the GCT discharges towards the area located southern the discharge source (~ 10 km; Fig. 5).

3.4. Assessment of the long-term effects of solid wastes on the central coastal area of GG

The central coastal area of GG has known various geomorphological changes due to the huge quantities of various solid industrial wastes (mainly PG) dumped by the GCT factories directly in the sea. In fact, just few years after the beginning of the GCT marine discharges, Darmoul (1988) observed a degradation of the *Posidonia oceanica* seagrass beds in front of the industrial emissary. Various authors highlighted that this degradation is mainly due to the effects of the untreated PG dumped in the coastal environment sea (Darmoul et al., 1980a; Darmoul, 1988; Zaouali, 1993; El Zrelli et al., 2017). This industrial waste was also reported to be responsible of other environmental consequences including the increase of water turbidity (Darmoul et al., 1980a), chemical contamination of seawater (Darmoul et al., 1980a; Kharroubi et al., 2012; El Zrelli et al., 2018b), destruction of the benthic community (Darmoul et al., 1980a and b; Rabaoui et al., 2014, 2015; El Kateb et al., 2016, 2018; El Zrelli et al., 2017) and silting up the seabed (Darmoul et al., 1980a; Zaouali, 1993). A recently-conducted study confirmed the cumulative negative impact of PG on *P. oceanica* meadows which disappeared -almost- completely from the study area, except for few small patches of this plant still remaining at almost 6 km (~ few hundred meters after the CM station described

herein; Fig. 1) to the south of the PG emissary (El Zrelli et al., 2017). Consequently, an azoic benthic zone was established in the coastal area between the two ports of Ghannouche and Gabes, inter-harbor sector (SM-Fig. 4; Rabaoui et al., 2015). In all, the local disappearance of *P. oceanica* seagrass can be considered as the major impact of GCT industrial marine discharges, in particular because this endemic plant represents a key species in the coastal ecosystem of GG (El Zrelli et al., 2017). Other serious changes caused by these industrial wastes took also place in the central coastal area of GG leading to more accentuate the degradation of the coastal habitat in this region. The surplus of industrial-derived coastal sediments have gradually led to a "fattening" of Chatt Essalam Beach (SM-Fig. 5) from the north to the south, in the direction of marine transport of industrial wastes (El Zrelli et al., 2015). This phenomenon was, firstly, caused the advancement of the shoreline more than 200 meters in the northern part of this area, close to the GCT industrial emissary (SM-Fig. 6), with the formation of a belt of some coastal dunes (SM-Fig. 7). The disappearance of an old coastal building of a red tuna processing plant present until 2013 (SM-Fig. 8), could be an additional proof of the beach flattening of the inter-harbor coastal area. In addition, the large quantities of industrial wastes have gradually clogging the mouths of 2 coastal streams (SM-Fig. 9) which has led to the formation of 2 small coastal lakes (Aouina and Ayatt lakes; SM-Fig. 10). It is worth noting that the coastal installations of commercial and fishing harbors might most likely play indirectly an essential role in the succession of these coastal modifications through the entrapment of a large proportion of these industrial wastes in the inter-harbor area. This can be confirmed with the Google Earth photo given in SM-Fig. 11, which showed that the sediments of the inter-harbor sector have a dark black color; whereas those of the southern area have a clear color.

The impacts of phosphate treatment industries on the coastal environment are known to differ with respect to hydrodynamic and geomorphologic features. For example, in the Lebanese coastal area of Batroun, the dumped PG discharges have affected the sediment quality leading to the prevalence of fine fractions, decreased pH and oxidation reduction potential, and increased inorganic phosphate concentrations (Fakhri et al., 2009, 2011). The latter authors have also observed a strong shrinkage of meiofauna representation and higher chlorophyll *a* concentrations in the sediments in the front of the PG emissary. Seawater quality was also found to be affected by PG wastes through the increase in its pH, temperature and orthophosphate concentrations (Fakhri et al., 2009, 2011). Similar impacts were also observed in the Seine Estuary (France), decades after banning the PG discharges in the Seine River

(Simon et al., 2000; Chiffoleau et al., 2001). This leads to deduce that the environmental impacts of PG releases observed in Gabes will most likely remain for decades, even if these industrial effluents are stopped, and that the restoration of the coastal environment which has been affected for decades (more than 50 years) will necessarily require a depollution phase of the water and sediments, followed by a monitoring program at different levels (fauna, flora, sediment, water…etc.). Moreover, the local regression/disappearance of *P. oceanica* seagrass from the central area of Gabes Gulf, caused by PG discharges, is most likely the triggering factor provoking, through a 'domino' effect, the other morphological impacts (SM-Fig. 4 to SM-Fig. 10). In fact, *P. oceanica* naturally ensures the stability of sandy shores and seabed (Maggi, 1973; Hattour and Ben Mustapha, 2013). Therefore, any future remediation program of the marine environment in Gabes should begin with the introduction of this endemic Mediterranean species which can help to restore the ecological balance of the GG ecosystem.

4. Conclusions

Summarizing, the present study showed that the solid industrial effluents are formed by phosphogypsum, fluorapatite, smokey quartz, calcite, fluorite, calcite-Mg, apatite, pyrite, albite, sphalerite-Cd-U, fluorosilicates, sulfur dross and spent catalysts. Essorrag Wadi represents the spatial limit of coastal sedimentary transport of the solid industrial wastes (phosphate grains). One must note that phosphogypsum was found to be completely dissolved in the seawater. Since the installation of the industrial complex in the coast of Gabes city in 1972, the coastal landscape has undergone several major modifications. The local extinction of *P. oceanica* seagrass, installation of azoic benthic seabed in the inter-harbor area, gradual fattening of beaches, advancement of the shoreline, formation of a costal dunes belt, clogging of the coastal streams' mouths and formation of coastal lakes represent the main coastal changes identified in this study. Based on these findings, a rapid intervention is necessarily and urgently needed in order to allow the recovery of the impacted coastal habitats of central Gabes Gulf. To ensure the success of any remediation plan of central GG, it is mandatory to ban dumping all types of industrial discharges in the marine environment. Achieving the restoration of Gabes Gulf environment cannot be done considering only this latter step. Other steps should be also considered including the decontamination of the seabed and clean-up of beaches. These measures will certainly help the re-apparition of locally-extinct taxa such as *P. oceanica* which play the role of a key species in the Gulf of Gabes.

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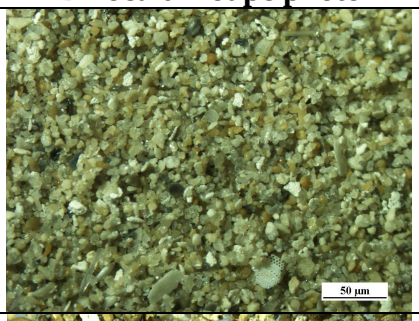
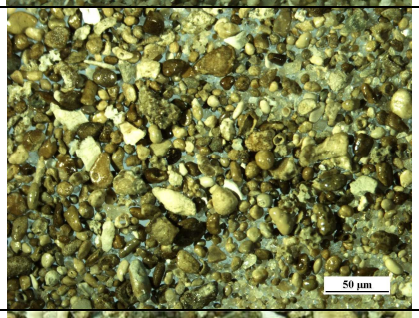
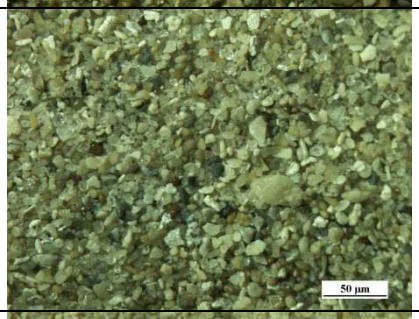
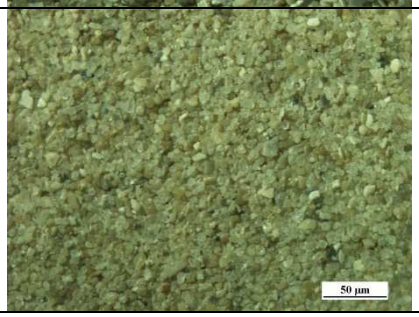
Table 1. Table summarizing the locations and number of trenches realized in each sampling station and in each sector, number and labels of sediments layers observed and number of samples collected from layers/trenches.

Sectors		Trenches		Latitude	Longitude	Layers	Number of samples
Inter-harbor Sector		LG		33.913183	10.103017	L4	2
		CE	CE1	33.900213	10.109032	L1, L2 and L3	3
			CE2	33.900336	10.109278	L1, L2 and L3	3
			CE3	33.900501	10.109706	L4	2
Southern Sector	Zone 1	CR		33.888367	10.119399	L1 and Ls	2
		OG		33.881070	10.123885		2
		CM		33.872555	10.131456		2
		MT		33.866872	10.136622		2
		TB		33.856182	10.147702		2
	Zone 2	LM		33.830594	10.175549	L1	1
		KT		33.791923	10.229626		1
		ZT		33.713848	10.326283		1

615 **Table 2**. Description of different coastal layers. (Ls: G×15; L4: G×10; L3: G×15; L2: G×20;
616 L1: G×20).
617

Layers	Binocular loupe photo	Description
L_s		**Color:** greyish **Composition:** Quartz, calcite, zircon, titanomagnetite, Calcite-Mg and small spherical shape fluorapatite grains (Ø<250µm), with a homogeneous fine granulometry. **Spatial extension:** in the southern area, from Corniche to Tebelbou (Sourrag Wadi).
L4		**Color:** Dark brown to blackish **Composition:** Almost dominance of fluorapatite grains, with the presence of smokey quartz grains, sphalerite-Cd, calcite-Mg, feldspar-K, fluorosilicates, sulfur dross and spent catalysts, with a very coarse heterogeneous granulometry. **Spatial extension:** in the inter-harbor area.
L3		**Color:** Brown **Composition:** Fluorapatite, quartz, calcite, zircon, calcite-Mg, sphalerite-Cd with the minority presence of barite and albite with heterogeneous medium granulometry. **Spatial extension:** in the inter-harbor area.
L2		**Color:** greyish **Composition:** Quartz, calcite, calcite-Mg, fluorapatite, zircon, fluorite, apatite and titanomagnetite, with a homogeneous medium to fine granulometry. **Spatial extension:** in the inter-harbor area.
L1		**Color:** white to blond yellowish **Composition:** Quartz grains, calcite grains (fragments of shells) and opaque grains (zircon, iron oxides and titanomagnetite minerals), with a homogeneous fine granulometry. **Spatial extension:** from Chatt Essalam to Zarat.

618

Figures Captions

Fig. 1. Location of the study area and sampling stations.

Fig. 2. Mineralogical composition of the sediment samples collected from the bed of the marine industrial discharges of the Gabes phosphate chemical complex (1: fluorapatite, 2: smokey Quartz, 3: calcite, 4: used catalysts, 5: sulfur dross, 6: sphalerite-Cd-U (+ fluorite), 7: apatite, 8: fluorosilicates. All the minerals presented here (1-8) were found in different samples and combined together just to show the mineralogical composition of these samples.

Fig. 3. SEM image and EDS spectra of Sphalerite-Cd-U.

Fig. 4. Sequential organization of L1, L2, L3, L4 and L_s layers at Gabes Beach. **A** and **B** at Chatt Essalam Beach, and **C** at M'torrech station (zone1).

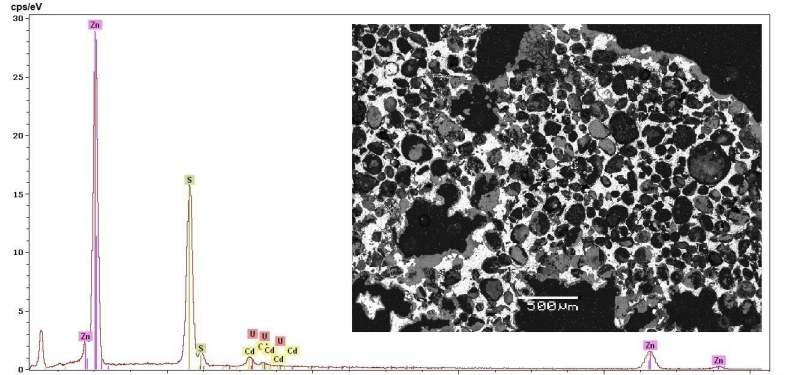
Fig. 5. North-South (N-S) sedimentary organization of the study beach area.

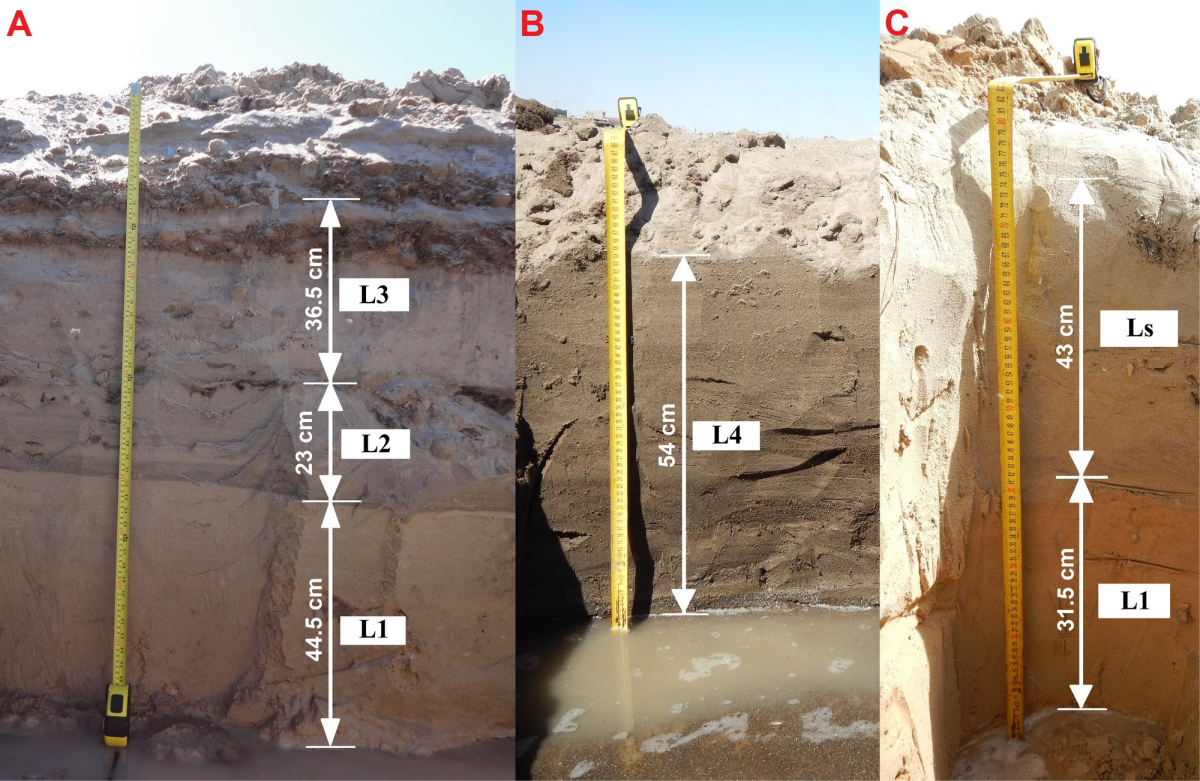
Fig. 6. Variations of Pb, Cd, Zn, Cr, Cu, Hg (in mg kg^{-1}) and P_{ing} (in g kg^{-1}) concentration (log-transformed) among the four different sediment layers observed and samples in Chat Essalam Beach.

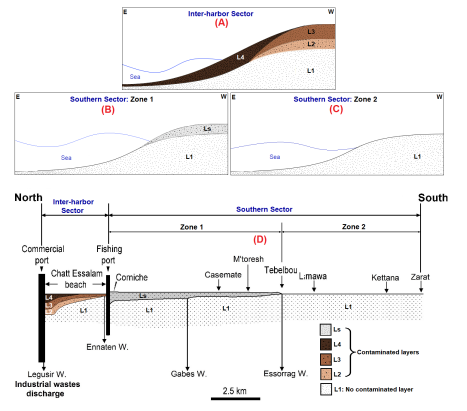
Fig. 7. SEM photos of (**A**) representative fluorapatite mineral grains from Legusir (LG), Chatt Essalam (CE) (Zone 1) and Tebelbou (TB) (Zone 2) stations and

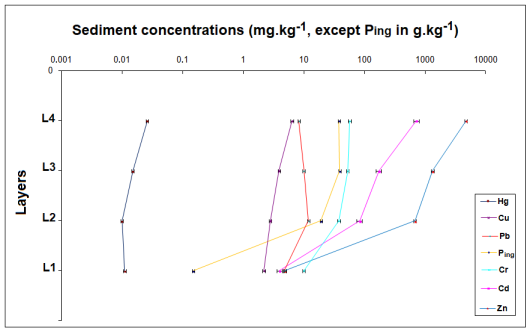

Industrial wastes discharge
LG
CE
Inter-harbor Sector
Gabes
CR
OG
CM
MT
TB
Essorag Wadi
Zone 1
Tebellbon
LM
Southern Sector
KT
Zone 2
Kettana
ZT
Zarat
0
5 Km
N
Italy
Ras Kaboudia
Tunisia
Mediterranean
Sea
Algeria
Libya
Gulf of Gabes

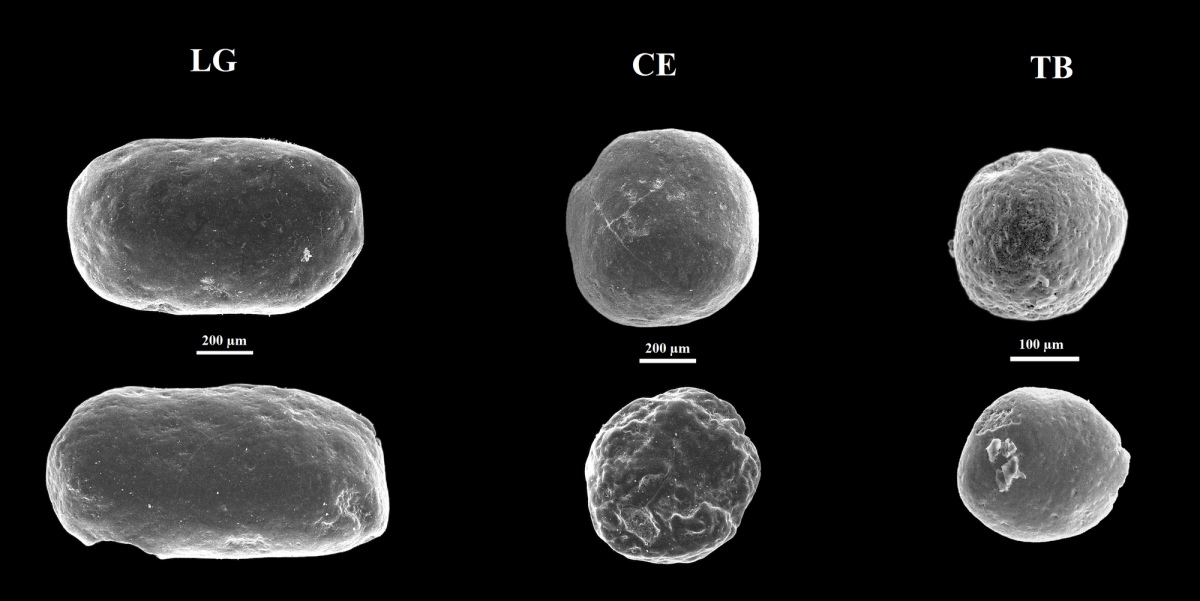

3
2
6
1
1
3
4
1
1
1
3
1
8
2
7
2
5
5
2
1
1
2
1 cm


cps/eV
30
25
20
15
10
5
0
Zn
Zn
S
S
U
U
U
Cd
Cd
Cd
Cd
Zn
Zn
2
4
6
8
10
KeV
500µm

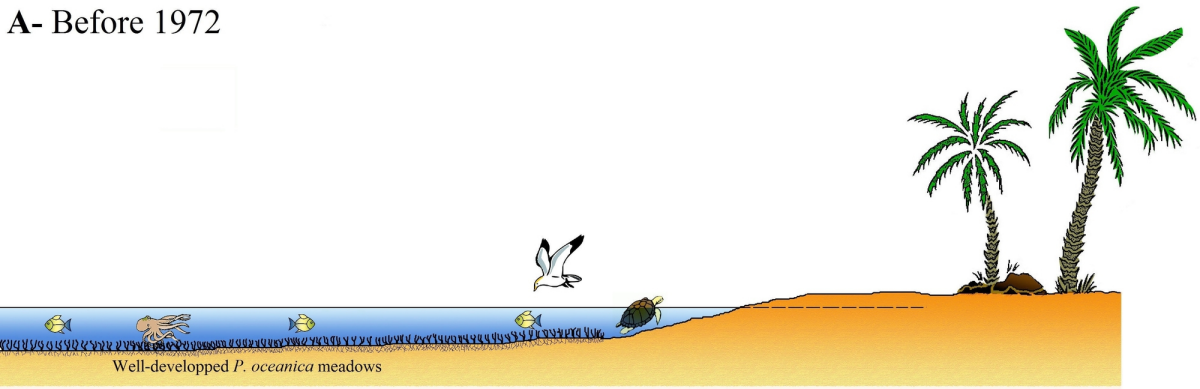

A
36.5 cm
L3
23 cm
L2
44.5 cm
L1
B
54 cm
L4
C
43 cm
Ls
31.5 cm
L1


E
W
Inter-harbor Sector
(A)
L3
L2
L4
Sea
L1
E
W
Southern Sector: Zone 1
(B)
L5
Sea
L1
E
W
Southern Sector: Zone 2
(C)
Sea
L1
North
South
Inter-harbor Sector
Southern Sector
Zone 1
Zone 2
(D)
Commercial port
Fishing port
Chatt Essalam beach
Corniche
Casemate
M'toreish
Tebelbou
Limawa
Kettana
Zarat
L5
L4
L3
L2
L1
Legsir W.
Industrial wastes discharge
Enhalen W.
Gabes W.
Essorrag W.
Contaminated layers
L1: No contaminated layer
2.5 km


Sediment concentrations (mg.kg-1, except Ping in g.kg-1)
0.001
0.01
0.1
1
10
100
1000
10000
0
Layers
L4
L3
L2
L1
Hg
Cu
Pb
Ping
Cr
Cd
Zn


LG
CE
TB
200 µm
200 µm
100 µm

A- Before 1972



B- After 1972

The main coastal changes caused by industrial solid wastes are: **l**ocal extinction of *Posidonia oceanica*, **i**nstallation of azoic benthic seabed in the inter-harbor area, **g**radual fattening of beaches, **a**dvancement of the shoreline, **c**logging of the coastal rivers mouths and **f**ormation of coastal lakes.